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Published in:
Proceedings of the Risø International Symposium on Materials Science

Publication date:
2012

Document Version
Early version, also known as pre-print

PREPARATION AND PROPERTIES OF TiB_2/TiAl COMPOSITE SHEETS WITH LAYERED STRUCTURE

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ABSTRACT

A novel TiB_2/TiAl matrix composite sheet with a layered structure has been successfully prepared by hot rolling of multi-layered Ti-(TiB_2/Al) sheets and subsequent multi-step heat treatment. TiB_2-rich layers can hinder the microstructural coarsening of the TiAl layer, with a temperature increase from room temperature to 700^{o}C, the tensile properties have been significantly improved caused by an increase of energy dissipation by plastic deformation and followed by a reduction in the sensitivity to micro crack formation.

1. INTRODUCTION

Gamma titanium aluminide (γ-TiAl) sheet has huge potential as a high-temperature structural material because of its excellent properties, such as high specific strength and modulus, low density (3.7 g·cm^{-3} ~ 4.1 g·cm^{-3}), high melting point and good resistance against oxidation and crrosion, especially for thermal protection systems in aerospace vehicles as a skin material (Koeppe, Bartels, Clemens, Schretter and Glatz, 1995; Gajanan, Chaudhari and Acoff, 2010). Nevertheless, the preparation of TiAl sheets is difficult due to its poor ductility and limited hot deformability, and a reproducible and controllable fabrication method of TiAl sheets for their practical application. As reported (Yang, Cizek, Hodgson and Wen, 2010; Luo and Acoff. 2006; Cao, Lin, Hu and Chen, 2008; Fukutomi, Ueno, Nakamura, Suzuki and Kikuchi, 1999), TiAl sheets can be manufactured by rolling pure Ti and Al foils together and subsequent reaction annealing. However, this method has several drawbacks (Cui, Fan, Geng, Wang, Zhang and Peng, 2012): i) it is rather difficult to obtain the dense TiAl sheets (Mackowiak and Shreir, 1959; Jakob and Speidel, 1994) due to a Kirkendall effect between Ti and Al, thus few studies on the mechanical properties of the TiAl sheets prepared by roll bonding and subsequent reaction annealing have been reported (Gajanan, Chaudhari and Acoff, 2010; Jakob and Speidel 1994); ii) the end-use temperature and mechanical properties of monolithic TiAl sheets cannot satisfy the requirement of aerospace application for high-temperature structural materials (Hodge, Hsiung and Nieh, 2004; Hazzledine and Kad, 2005).

In the present work, TiB_2/Al composite sheets are applied to replace aluminum sheets, whilst an effective densification treatment is utilized to increase the relative density of multi-layered TiB_2-TiAl composite sheets which combine the respective advantages of intermetallic TiAl matrix and ceramic TiB_2 particles (Wang, Lin, He, Wang and Chen, 2009). As a result, owing to nanoscale spacing of γ-TiAl/α_2-Ti_3Al layers and a multi-interface effect of the special layered structure, such as thermal stability of the microstructure (Ding, Northwood and Alpas, 1993; Rowe, Skelly, Larsen, Heathcote, Odette and Lucas, 1994), the sheets exhibit improved room temperature and high temperature tensile strength compared with monolithic TiAl.

2. EXPERIMENTAL PROCEDURES

Thin commercial pure Ti sheets with a thickness of 200μm and 2 vol.% TiB_2/Al (Al matrix with a 99.5% purity) composite sheets with a thickness of 250 μm were cut into 50 mm × 50 mm squares. Surface pre-treated Ti and TiB_2/Al sheets were alternately stacked to form a “sandwich” and wrapped with Al foil and hot-rolled at 510 ^{o}C with a 60% reduction in thickness. The hot-rolled laminates were annealed in a vacuum furnace as follows: i) annealing at 650 ^{o}C for 40h in a stainless steel mould fixed with screws and densifying in vacuum under 50 MPa at 1225 ^{o}C for 2h to eliminate the Kirkendall voids formed during the initial annealing process; ii) second annealing for composition homogenization at 1250 ^{o}C for 15h ; iii) final annealing at 1400 ^{o}C for 20 minutes in order to obtain an alternating lamellar structure of TiAl and Ti_3Al. The final thickness of the multi-layered composite sheets is in the range of 2.1 – 2.3 mm.

The cross sections of samples (Table 1) following the above annealing processes were examined with a scanning electron microscope (SEM, FEI Quanta 200F) equipped with an energy dispersive X-ray spectrometer (EDX). In addition, a transmission electron microscope (TEM, FEI Tecnai F30) was used to observe the microstructure and determine the different phases. All examinations were taken in the ND-TD plane of the sheets.

The tensile samples were cut parallel to the rolling direction using electrical discharge machining (EDM) and their surfaces were carefully polished to remove the machining traces. The cross-section of tensile samples was 2.5 mm × 1.5 mm and the gauge length was 15mm. The tensile tests were carried out at room temperature and 700^{o}C with an Instron-1186 universal testing machine with an initial strain rate of 1.3×10^{-4} s^{-1}. The density of the composite was measured to be 3.8 $g\cdot cm^{-3}$ by Archimedes method.

Table 1 Information of samples

Samples	Sample 1 (Fig. 1)	Sample 2 (Fig. 2(a))	Sample 3 (Fig. 2(b))	Sample 4 (Fig. 3)
Processing	After hot rolling at 510 ^{o}C	After first annealing at 650 ^{o}C for 40h	After second annealing at 1250 ^{o}C for 15h	After final annealing at 1400 ^{o}C for 20 minutes

3. RESULTS AND DISCUSSION

3.1. Hot rolling. The microstructure of the hot-rolled sample is shown in Fig. 1. Fig. 1(a) shows that the deformation of Ti and TiB_2/Al composite layer is uniform, only local necking of the Ti layer has been observed. As shown in Fig. 1(b), the interfacial bonding between Ti and the TiB_2/Al composite layer is compact and no intermetallic compounds have formed.

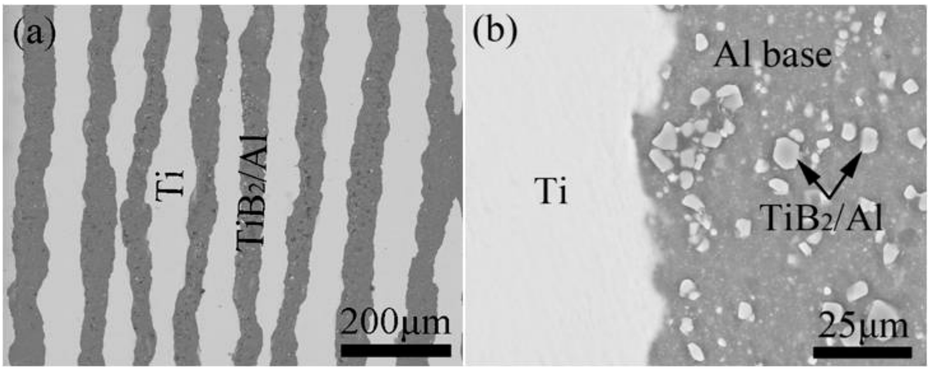

Fig. 1. Backscattered electron (BSE) images of (a) the cross section of hot-rolled sample and (b) a magnified zone of the interface of Ti and TiB_2/Al layers.

3.2. Initial annealing, densification process and second annealing. After hot rolling, the multi-layered Ti-(TiB_2/Al) sheets were annealed in a vacuum furnace. The initial annealing temperature was (650^{o}C) selected slightly below the melting point of pure Al (660^{o}C) for (i) assuring the high reaction-rate of Ti and Al; (ii) prohibiting the leaking of Al. Fig. 2(a) shows that $TiAl_3$ is the only intermetallic phase. SEM/EDX results show that Al has been exhausted and residual Ti layers and unreacted TiB_2 are observed after the initial annealing (650 ^{o}C for 40h). During the initial annealing process, numerous pores formed in the original TiB_2/Al composite layer due to the Kirkendall effect, which would reduce the mechanical properties of the laminates. Therefore, a hot pressing at 1225 ^{o}C for 2h under 50 MPa was used to eliminate the pores within the laminates and to improve the density. In addition, the hot pressing decreased the layer thicknesses, which would shorten the second annealing time.

After the densification treatment, the laminates were heat-treated by the second annealing at 1250 ^{o}C for 15h. As shown in Fig. 2 (b), the results of SEM/EDX show that pure Ti and $TiAl_3$ layers have disappeared, and alternating TiB_2, TiAl and Ti_3Al layers can be detected. The inset of Fig. 2 (b) indicates that the TiB_2 layer contains a low volume fraction of TiAl (labeled as TiB_2-rich layer) is compact.

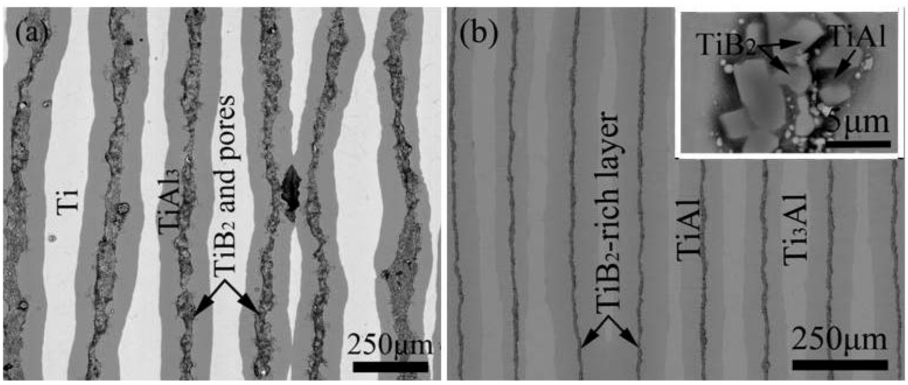

Fig. 2. Microstructure of the multi-layered Ti-(TiB_2/Al) sheets treated by (a) first annealing at 650 ^oC for 40h and (b) second annealing at 1250 ^oC for 15h after a expanded densification treatment at 1225 ^oC for 2h under 50 MPa.

3.3. Homogenization treatment. To homogenize the microstructure and obtain the lamellar γ-TiAl/α_2-Ti_3Al structure, the homogenization treatment was conducted at 1400 ^oC for 20 minutes. As shown in Fig. 3(a), voids have disappeared and the dense multi-layered microstructure consists of alternating TiB_2-rich layers with a thickness of approximately 5μm and γ-TiAl/α_2-Ti_3Al layers (with several colonies of lamellar γ-TiAl/α_2-Ti_3Al structure) with an average width of 160 μm. the detailed microstructural features of the composite are sketched in Fig. 3(b). From the TEM image of the TiB_2-rich layer (Fig. 3(c)), the layer consists of TiB_2 particles and γ-TiAl, and no voids can be found. As shown in Fig. 3(d), the colony of the lamellar γ-TiAl/α_2-Ti_3Al structure is composed of γ-TiAl and α_2-Ti_3Al lamellae (the average spacing ≈ 100 nm) with the orientation relationship: $[11\text{-}20]_{\alpha 2}//[110]_\gamma$ and $(0001)_{\alpha 2}//(1\text{-}11)_\gamma$. The thickness ratio of TiB_2 and γ-TiAl/α_2-Ti_3Al layers can be controlled by a variation in the volume fraction of the original TiB_2, Al and Ti sheets. The colony size range of the lamellar γ-TiAl/α_2-Ti_3Al structure is 50 ~ 160μm and the average size is 90μm determined by the line intercept method, which is smaller compared with the γ-TiAl alloys produced by other methods (Cao, Lin, Hu and Chen, 2008). This is because the dense TiB_2-rich layer significantly restricts the coarsening of the colony of the lamellar γ-TiAl/α_2-Ti_3Al structure. The EDS result shows that, the composition of the multi-layered sheets is homogeneous. The Ti contents in the present work lie in the stability range of the (α_2+γ) and γ phase regions, which corresponds to the lamellar γ-TiAl/α_2-Ti_3Al structure.

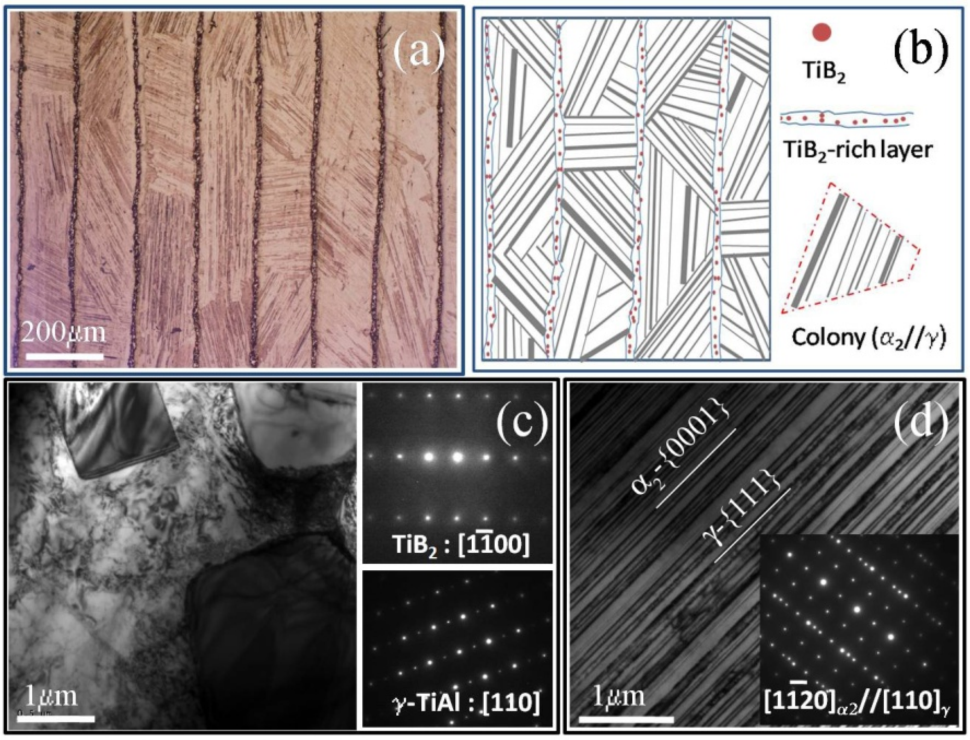

Fig. 3. (a) Optical image of the multi-layered TiB_2-TiAl composite sheets after the homogenization treatment and (b) a corresponding schematic illustration of the layered structure of TiB_2-rich layers and the lamellar γ-TiAl/α_2-Ti_3Al structure, TEM images of (c) TiB_2-rich layer and (d) the lamellar γ-TiAl/α_2-Ti_3Al structure imaged in the $[110]_\gamma$ and $[11\text{-}20]_{\alpha 2}$ direction.

<u>3.4. Tensile properties</u>. Fig. 4(a) shows the tensile properties of the multi-layered TiB_2-TiAl composite sheets at room temperature and 700 ^oC. The ultimate tensile strength (σ_b), yield strength ($\sigma_{0.2}$) and elongation (δ) of the sheets at ambient temperature are 215.6 MPa, 201.5 MPa and 1.31 %, respectively. The overall fracture surface of the composite at room temperature (Fig. 4(b)) is very flat and has some large cleavage faces, indicating a typical brittle failure. The reason why the multi-layered composite sheets exhibits low room-temperature tensile properties is that the TiB_2-rich layers are rather brittle at room temperature and that a small amount of pores may still be present within the TiB_2-rich layers where micro-cracks can form and propagate easily. As a result, a fracture process can be rapid and the strength of the multi-layered composite sheets cannot be fully exploited. However, the composite sheets show excellent high-temperature properties. At 700 ^oC, the σ_b, $\sigma_{0.2}$ and δ rise to 312.6 MPa, 300.8 MPa and 3.08 %, respectively, which are higher than those of monolithic TiAl prepared by roll bonding and reaction annealing (Jakob and Speidel, 1994).

As the testing temperature increases, the elongation shows a sharp increase (Fig. 4(a)) owing to the activation of additional slip systems, which approaches the value of monolithic TiAl prepared by roll bonding and reaction annealing (Jakob and Speidel, 1994). Fig. 4(c) shows the fracture surface of the specimen tested at 700 ^oC, which demonstrates the tearing-out deformation structure within the lamellar colony, while the fracture surface at ambient temperature is nearly free of a deformation structure. The room-temperature toughness of TiAl-based materials is sensitive to microstructure, e.g. pores, and the cleavage fracture occurs generally on low-index crystallographic planes. A high temperature tends to decrease the yield points and to enhance plastic deformation. Thus, during crack propagation the energy dissipation by plastic deformation becomes more important, the higher the test temperature (Appel, Paul and Oehring, 2011). In the multi-layered TiB_2-(γ-TiAl/α_2-Ti_3Al) composite, at a higher temperature dislocations can glide and climb due to thermal activation. Deformation processes can easily spread within the plastic zone of the cracks so that the constraints due to the local slip geometry are less restrictive. Therefore, the fracture mechanism changes from a cleavage-dominated fracture at ambient temperature to an energy-absorbing ductile fracture at high temperature. A detailed investigation of the dislocation activity during the high-temperature deformation is a part of on-going research.

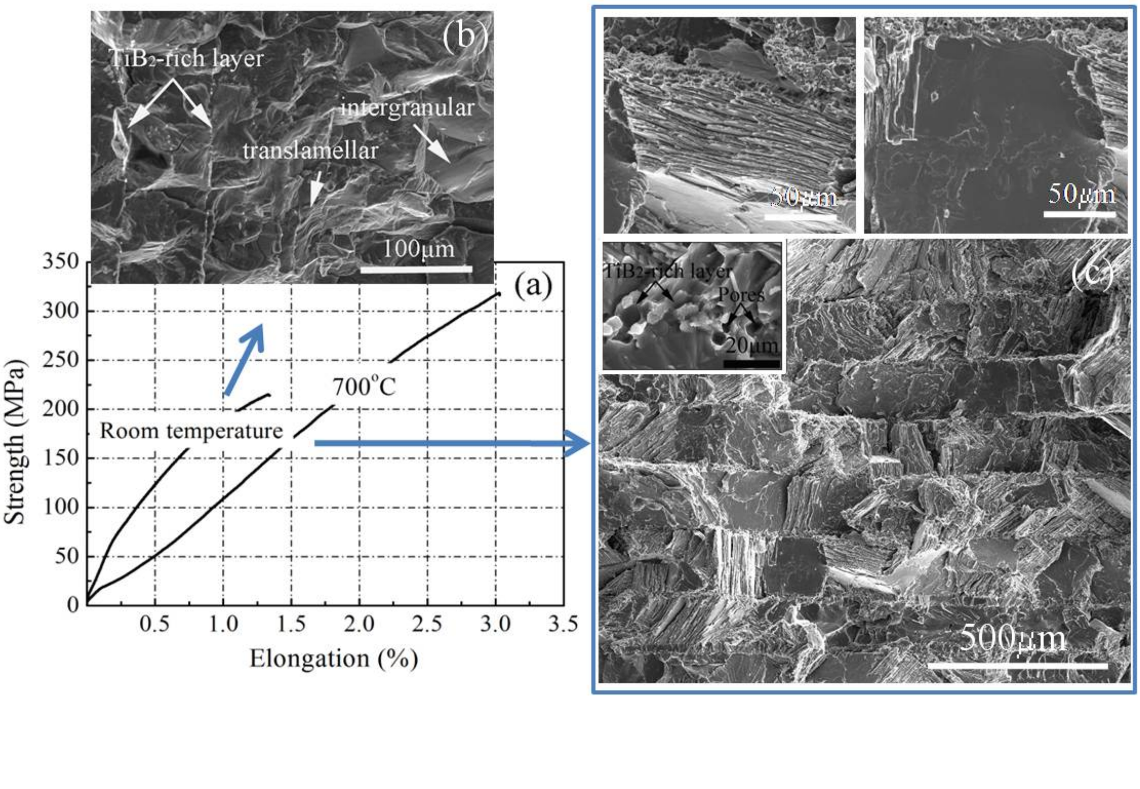

Fig. 4. (a) tensile properties of the multi-layered TiB_2-(γ-TiAl/α_2-Ti_3Al) composite sheets; and the fracture surfaces of samples tested at (b) room temperature and (c) 700 ^oC with the tearing-out feature.

4. CONCLUSIONS

1. A dense multi-layered TiB_2-TiAl composite structure can be produced by hot rolling and reaction annealing of stacks of alternating layers of pure Ti and 2 vol.% TiB_2/Al.

2. The thickness ratio of TiB_2-rich and γ-TiAl/α_2-Ti_3Al layers in the composite can be controlled by variation in the initial volume contribution of TiB_2, Al and Ti.

3. The composite is fairly brittle when tested at room temperature but both the strength and elongation is significantly improved when tested at 700 °C. The composite therefore has potential for high temperature application as a strong and light material.

ACKNOWLEDGEMENTS

This work was financially supported by (a) the National Natural Science Foundation of China (Project No.: 51071058, 51101041) and (b) the Fundamental Research Funds for the Central University (Contract No.: HIT. ICRST.2010007, HIT. NSRIF. 2010107). The authors thank Dr. N. Hansen for his useful comments during the preparation of the manuscript.

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